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AF/TFW

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

Appl. No. : 10/543,121 Confirmation No. 4365
Applicant(s) : Gilbert Bouquet, et al
Filed : July 22, 2005
TC/A.U. : 1796
Examiner : Olga Asinovsky
Title : PARTICLE SIZE AND MORPHOLOGY CONTROL IN RUBBER
MODIFIED MONOVINYLDENE AROMATIC POLYMERS
Docket No. : 62723A
Customer No. : 00109

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APPEAL BRIEF

Responsive to the Official Action dated January 25, 2008.

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Real Party in Interest

The subject application is owned by Dow Global Technologies, Inc.

Related Appeals and Interferences

At the time there are no related appeals or interferences.

Status of the Claims

On April 24, 2008, Appellant appealed the final rejection of Claims 1 to 13 and 17 to 19, Claims 14 to 16 having been canceled.

Status of the Amendments

No amendments have been submitted subsequent to the final rejection dated January 25, 2008.

Summary of Claimed Subject Matter

As described in more detail in the application, Appellant's invention (independent Claim 1) is:

- page 3, lines 6-9 a solution for achieving rubber particle size, distribution and morphology control in the production of rubber modified polymers at an increased rate
wherein
- page 3, lines 10-13 the process is a mass/solution polymerization process utilizing a functionalized rubber to produce a rubber modified polymer from a vinyl aromatic monomer comprising polymerizing the vinyl aromatic monomer in the presence of a rubber
wherein
- page 3, line 13 the rubber comprises a functionalized diene rubber having:
- page 3, line 14 and a solution viscosity of from 5 to less than 50 centipoise
page 4, lines 4-6 and
page 3, line 15 and at least one functional group per rubber molecule which
page 5, lines 4-5 enables controlled radical polymerization
page 3, lines 16-17 such that grafted rubber particles are formed and dispersed within a matrix comprising polymerized vinyl aromatic monomer and have a broad monomodal size distribution.

Grounds of Rejection to be Reviewed on Appeal

Whether Claims 1 to 13 and 17 to 19 are unpatentable under 35 U.S.C. 103(a) over US 5,721,320 (Priddy) in view of EP 0892820.

Argument

Rejection under 35 U.S.C. 103(a) over Priddy in view of EP 0892820:

Claims 1 to 13 and 17 to 19

The Examiner has rejected Claims 1 to 13 and 17 to 19 under 35 U.S.C. 103(a) as being unpatentable over Priddy et al (USP 5,721,320) in view of EP 0892820.

As described in more detail in the application, Applicant claims a mass/solution polymerization process comprising polymerizing a vinyl aromatic monomer in the presence of a functionalized diene rubber having a solution viscosity of from 5 to less than 50 centipoise wherein the functional group on the rubber enables controlled radical polymerization such that the resulting grafted rubber particles have a broad monomodal size distribution.

Priddy discloses a bulk (mass) polymerization process comprising a rubber carrying a functionalized group enabling controlled radical polymerization with a vinyl aromatic monomer, preferably to prepare a transparent rubber-modified polymer. Priddy is silent as to viscosity of the functionalized rubber and to the rubber particle size distribution.

EP 0892820 discloses a process for preparing modified polymers from vinyl aromatic monomers wherein the rubber has a bimodal particle size distribution. EP 0892820 is silent as to the viscosity of the rubbers used in its process.

Neither Priddy nor EP 0892820 teach or suggest any viscosity limitations regarding the rubbers used in their respective inventions. Applicant asserts that it is impossible for one skilled in the art to combine Priddy with EP 0892820 to arrive at Applicant's invention of a polymerization process comprising polymerizing a vinyl aromatic monomer in the presence of a functionalized diene rubber having a solution viscosity of from 5 to less than 50 centipoise (cps) because neither disclose anything about rubber solution viscosity.

While neither Priddy nor EP 0892820 teach or suggest any viscosity limitations regarding the rubbers used in their respective inventions, the Examiner cites Example 1 wherein a functionalized rubber having a Mw of 3930 is reacted with styrene to provide a transparent rubber-modified polystyrene (OA dated August 8, 2007). The Examiner concludes that it would be obvious to arrive at Applicant's

claimed viscosity range from the low molecular weight of the functionalized polybutadiene of Priddy. Applicant respectfully disagrees because one skilled in the art knows that solution viscosity is dependent on several variables, not just polymer molecular weight. As can be seen by Figure 1, of the Affidavit provided by Gilbert C. Bouquet, a relationship between Mw and solution viscosity for butadiene rubbers is graphically represented. The Mw/solution viscosity data points are from (1) the rubbers from the Examples of the present invention, (2) JP 4-88006 (Table 1) and (3) as determined by Dow for two commercially available butadiene block rubbers. As can be seen from the plot in Figure 1, a rubber with a Mw of 3930 (such as the rubber oligomer disclosed in Priddy) would have a solution viscosity significantly less than 5 cps. Priddy's low Mw rubber oligomer having a Mw of 3930 is clearly outside of the claimed invention.

Further, one skilled in the art knows that lower rubber solution viscosity (i.e., lower Mw as shown in Figure 1) is one (but not only) approach to producing smaller rubber particle size (see solution viscosity versus rubber particle size discussion hereinbelow) and that smaller rubber particle size is key in achieving transparency in rubber-modified polymers. Priddy's examples to make a transparent polymers using ultra low Mw rubber oligomers teaches away from Applicant's rubber which would have a much higher Mw as evidenced by the relationship in Figure 1 and having a solution viscosity range of from 5 to less than 50.

Priddy discloses rubbers with a Mw of between 20,000 to 300,000, most preferably 100,000 to 150,000. However, one can not rely solely on rubber Mw to predict its solution viscosity. The viscosity of a rubber is a function of more than just its molecular weight (for example whether it is a homopolymer, a copolymer (e.g., butadiene and styrene), if it is a copolymer how much copolymer is present (percent styrene), if the rubber polymer is linear or branched, etc.) one skilled in the art can not predict whether the molecular weight rubbers of Priddy will fall within the Applicant's selected range.

For example, in Response A, Gilbert C. Bouquet provided the following data for two functionalized rubbers, one (Rubber 1) with viscosity within the range of the present invention and the other (Rubber 2) having a viscosity outside the range of the present invention. However, Rubber 1 having a lower viscosity has a higher Mw than Rubber 2:

<u>Rubber</u>	<u>Mw, g/mole</u>	<u>Viscosity, cps</u>
1	192,800	33
2	168,900	51

As can be seen, Rubber 2, a rubber having a Mw within the most preferred Mw range of Priddy does not fall within the claimed range of solution viscosity in the present invention. Priddy and EP 0892820 are silent regarding rubber solution viscosity. Further, Priddy discloses rubber Mw ranges, but EP 0892820 is silent as to rubber Mw. Moreover, neither Priddy nor EP 0892920 teach or disclose any relationship between rubber Mw and solution viscosity which could enable one skilled in the art to predict that (1) a rubber with a certain Mw will have a certain viscosity based on Mw data alone nor (2) arrive at Applicant's solution viscosity range selection of from 5 to less than 50 cps.

To support that Applicant's assertion that the claimed selected range of solution viscosity of 5 to less than 50 cps for the functionalized rubber of the present invention is inventive, Applicant refers to Comparative Example A in the Affidavit by Gilbert Bouquet. Comparative Example A is prepared by the same process as Examples 1 to 3 of the present invention. The functionality on the rubber in Comparative Example A is the same functionality as on the rubbers in Examples 1 to 3. The process of Comparative Example A is not an example of the present invention because the solution viscosity of the functionalized rubber is greater than 50 cps. The following table compares the superior higher gloss of the product from the process of Examples 1 to 3 to the lower gloss of the product from the process of Comparative Example A:

Example Com. Ex.	Initiator top R1 (ppm)	NDM top R1 (ppm)	NDM top R2 (ppm)	RPS (μ m)	Intr.Glos (%)
1	120	300	1000	0.45	82
	120	200	1000	0.39	88
	120	100	1000	0.42	84
	120	150	1000	0.38	87
	120	250	1000	0.39	90
	110	200	1000	0.39	89
	100	200	1000	0.40	89
2	100	200	1000	0.38	86
	100	150	1000	0.39	87
	100	250	1000	0.41	88
	100	100	1000	0.40	88

	100	50	1000	0.39	89
	90	50	1000	0.41	88
	90	0	1000	0.41	88
3	90	0	1000	0.40	87
	90	50	1000	0.41	87
	90	100	1000	0.40	87
	90	150	1000	0.42	86
	90	200	1000	0.42	84
	100	200	1000	0.41	85
	100	250	1000	0.46	82
A*	100	250	1000	0.61	67
	100	250	1200	0.63	64
	90	250	1200	0.66	66
	90	200	1200	0.59	69
	90	150	1600	0.75	61

*not an example of the present invention

The Examiner makes a further argument that smaller rubber particle size improves gloss and that Priddy disclose rubbers having a particular rubber size (less than 0.1 micron) and that EP 0892820 also discloses rubbers of a similar particle size (OS dated January 25, 2008). The Examiner further states that EP 0892820 teaches smaller rubber particle size improve gloss (higher gloss value). It is well known in the art that smaller rubber particle size can improve gloss.

The Examiner seems to be suggesting that the only factor effecting rubber particle size is rubber solution viscosity; one skilled in the art knows this is not the case. This is even addressed in EP 089280 in paragraph 27:

"The particle size of the rubber particles produced in the first and second prepolymer compositions can be influenced by a number of factors including the rubber used, the amount of grafting, the viscosity and the shear rate. The techniques of mass-polymerization and the conditions needed for producing the desired average particle sizes are well known to one skilled in the art."

Wherein the viscosity mentioned in paragraph 27 could be either or both the viscosity of the matrix polymer and/or the rubber.

The Examiner seems to be suggesting that a specific rubber solution viscosity is an inherent property of a specific rubber particle size; however, one skilled in the art knows that it is not the case. It is well known that rubber viscosity depends on as many, if not more, variables than rubber particle size, i.e., type of rubber, homopolymer, copolymer, types of comonomers, block, random, molecular weight, cross linking, morphology, etc. There is no universal correlation between rubber particle size and rubber solution viscosity. Applicant

asserts that it is impossible for one skilled in the art to combine the teachings of Priddy with EP 0892820 in respect to rubber particle size to arrive at Applicant's invention of a polymerization process comprising polymerizing a vinyl aromatic monomer in the presence of a functionalized diene rubber having a solution viscosity of from 5 to less than 50 cps.

Priddy is silent as to the rubber particle size distribution in the products of its process (e.g., monomodal, bimodal, etc.). The composition disclosed in EP 0892820 has a bimodal rubber particle size distribution. One skilled in the art could not combine Priddy and EP 0892820 to arrive at Applicant's mass/solution polymerization process which provides a rubber modified polymer comprising grafted rubber particles having a broad monomodal size distribution.

One skilled in the art could not interpret the cited prior art to arrive at the Examiner's conclusions that Priddy combined with EP 0892820 teaches Applicant's process utilizing a functionalized rubber having a solution viscosity of from 5 to less than 50 cps and providing a rubber modified polymer comprising grafted rubber particles having a broad monomodal size distribution.

Applicant has shown that:

- (1) Applicant's invention is a selection of a range of functionalized rubbers providing unexpected and improved properties.
- (2) Priddy's rubber with a molecular weight of 3,930 is outside of Applicant's claimed range (low side) of a functionalized rubber with a solution viscosity of from 5 to less than 50 cps.
- (3) Rubbers having a molecular weight within the range disclosed in Priddy but having a solution viscosity outside of the Applicant's claimed range (high side) do not provide the unexpected improvement in gloss in the resultant rubber modified polymers compared to rubber modified polymers made by the process of the present invention having a solution viscosity within the claimed range.
- (4) Neither Priddy nor EP 0892820 teach or suggest Applicant's claimed functionalized rubber having a solution viscosity of from 5 to less than 50 cps.

(5) Neither Priddy nor EP 0892820 teach or suggest Applicant's claimed rubber particles having a broad monomodal size distribution.

Applicant asserts that the Examiner has not establishes a case of prima facie obviousness and that the 103(a) rejections are unsupported by the cited prior art. The Examiner has ignored two material limitations of the present invention, namely (1) that the functionalized rubber must have a solution viscosity of from 5 to less than 50 cps and (2) that the rubber particles have a broad monomodal size distribution. Both cited references are silent as to rubber solution viscosity. One reference is silent as to the particle size distribution of the rubber while the other discloses a bimodal rubber particle size distribution. The Examiner has not established a case of prima facie obviousness because the references cited do not contain every feature of the claimed invention. Applicant asserts that the present invention as claimed in present Claims 1 to 13 and 17 to 19 is inventive and patentable over Priddy in view of EP 0892820.

Conclusion

For the foregoing reasons, it is submitted that the Examiner's rejections of Claims 1 to 13 and 17 to 19 were erroneous and reversal of his decisions is respectfully requested.

Respectfully submitted,



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Claims Appendix

1. (previously amended) A mass/solution polymerization process utilizing a functionalized rubber to produce a rubber modified polymer from a vinyl aromatic monomer comprising polymerizing the vinyl aromatic monomer in the presence of a rubber, wherein the rubber comprises a functionalized diene rubber having:
 - a) a solution viscosity of from 5 to less than 50 centipoise (cps), and
 - b) at least one functional group per rubber molecule which enables controlled radical polymerization;such that grafted rubber particles are formed and dispersed within a matrix comprising polymerized vinyl aromatic monomer and have a broad monomodal size distribution.
2. (original) The process of Claim 1 wherein the vinyl aromatic monomer is styrene.
3. (original) The process of Claim 1 wherein the vinyl aromatic monomer is copolymerized with acrylonitrile.
4. (original) The process of Claim 1 wherein the functionalized diene rubber is a styrene/butadiene block copolymer rubber.
5. (original) The process of Claim 1 wherein the functionalized diene rubber has a solution viscosity of 5 weight percent in styrene at 20°C of less than 45 cps.
6. (previously amended) The process of Claim 1 wherein the functionalized diene rubber contains a functional group which enables controlled radical polymerization through formation of a stable free radical.
7. (original) The process of Claim 6 wherein the functionalized diene rubber contains a nitroxide functional group.
8. (original) The process of Claim 6 wherein the functionalized diene rubber contains a functional group selected from 2,2,6,6,-tetramethyl-1-piperidinyloxy (TEMPO); 2,2,6,6-tetramethyl-1-[1-[4-(oxiranylmethoxy)phenyl]ethoxy]-piperidine; or 3,3,8,8,10,10-hexamethyl-9-[1-[4-(oxiranylmethoxy)phenyl]ethoxy]-1,5-dioxa-9-azaspiro[5.5]undecane.
9. (previously amended) The process of Claim 1 wherein the functionalized diene rubber contains a functional group which enables controlled radical polymerization through atom transfer radical polymerization.

10. (previously amended) The process of Claim 1 wherein the functionalized diene rubber contains a functional group which enables controlled radical polymerization through reversible addition-fragmentation chain transfer polymerization.
11. (original) The process of Claim 1 wherein the polymerization is conducted in the presence of a chain transfer agent.
12. (original) The process of claim 1 wherein the polymerization is conducted in the presence of an initiator.
13. (original) The process of claim 1 wherein the polymerization is conduction in the presence of an initiator and a chain transfer agent.
14. (canceled)
15. (canceled)
16. (canceled)
17. (original) The process of claim 1 wherein a portion of a partially polymerized feed is recirculated to an earlier polymerization stage.
18. (original) The rubber modified polymer produced by the process of Claim 1.
19. (original) An article or composition comprising the rubber modified polymer of Claim 18.

Evidence Appendix

Starting on the next page (page 19) is a copy of the Affidavit under 37 CFR 1.132 and a copy of a Subscribing Witness Affidavit of Execution form both filed November 5, 2007, which was entered by the Examiner in her Office Action dated January 25, 2008.



UNITED STATES PATENT AND TRADEMARK OFFICE

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Melissa Van Meter

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November 1, 2007

DATE OF SIGNATURE

AFFIDAVIT Under
37 CFR 1.132

STATE OF MICHIGAN)
) ss.
County of Midland)

I, Gilbert C. Bouquet declare and state:

I am a citizen of Belgium, and reside at Gent, Estafetteweg 14.

I received a Bachelor Degree in Chemistry from The University of Gent in 1980; that I received a Master degree in Chemistry from The University of Gent in 1982; and that I received the degree of Doctor of Philosophy in Polymer Chemistry from The University of Gent in 1988.

I joined Dow Benelux NV in Terneuzen, Netherlands as a Research Chemist in Terneuzen in 1987, and currently I am a Senior Research Specialist within the Performance Plastics Business of The Dow Chemical Company;

I have 18 years of experience with acrylonitrile, butadiene, and styrene terpolymers (ABS), including (i) their composition, (ii) their methods of preparation, (iii) their use in various articles of manufacture, and (iv) the measurement and interpretation of their various properties.

I have presented herein below a correlation between molecular weight (Mw) and solution viscosity for butadiene rubbers used in the Examples of the present invention, values reported in the prior art, and values determined by Dow.

Solution Viscosity versus Molecular Weight For Diene Rubbers

The molecular weights and solution viscosities for the rubbers used in the Examples of US Application Serial No. 10/543,121 are as follows:

Example	Rubber	Mw (kg/mole)	Solution Viscosity (cps)
1	A	93000	10
2	B	135000	25
3	C	193000	33
4	E	115000	24
5	F	90000	10

The molecular weights and solution viscosities for the rubbers reported in JP 4-88006 (Table 1) and determined by Dow Chemical on two commercial rubbers is as follows:

Mw	Solution Viscosity (cps) wt% Styrene	
Reported in JP 4-88006:		
425000	65	30
435000	62	35
410000	68	25
400000	60	29
400000	60	29
460000	90	30
320000	57	25
400000	60	29
350000	33	50
400000	60	29
230000	33	30
520000	105	30
400000	60	29
400000	60	29

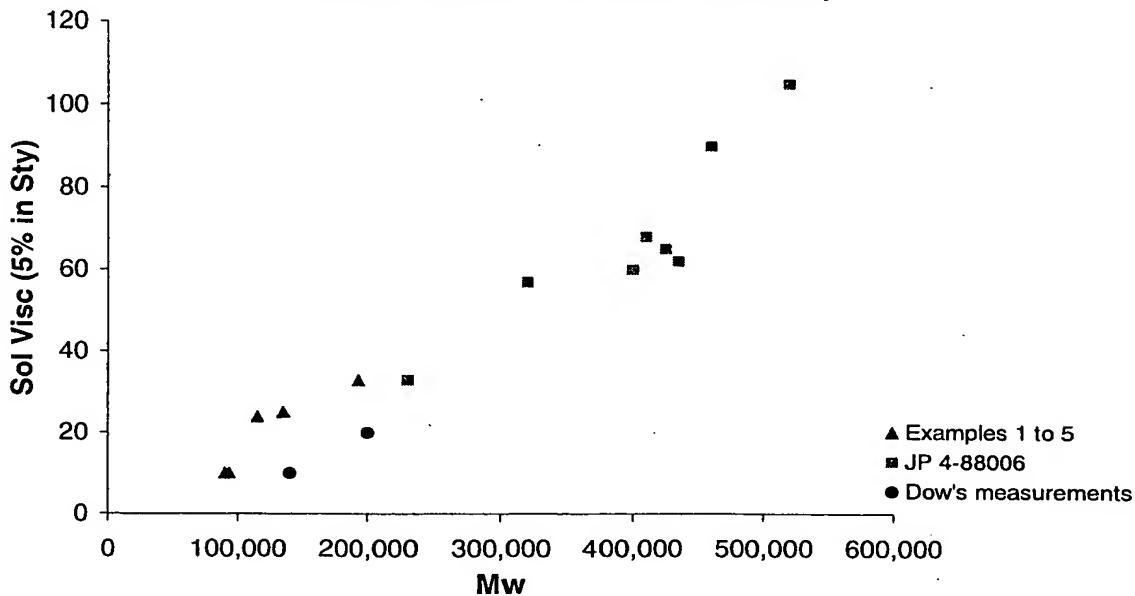
Dow analysis of commercial rubbers:

STEREON 205	140000	10	30
JT-42	200000	20	5

STEREONTM 205 is a 25wt% styrene/75wt% butadiene block rubber commercially available from Firestone with a Mw of 140,000 and JT-42 is a 5wt% styrene/95wt% butadiene block rubber commercially available from Asahi with a Mw of 200,000 kg/mole.

The data is represented graphically below in Figure 1:

FIG. 1
Rubber Mw v. Solution Viscosity



Further, I have conducted and supervised the following test in order to determine, and evaluate the resulting properties, especially gloss, of an ABS resin manufactured from a functionalized rubber having a specific solution viscosity.

Comparative Example A

A continuous polymerization apparatus composed of three 2.4 liter plug flow reactors connected in series, wherein each plug flow reactor is divided in three zones of equal size, each zone having a separate temperature control and equipped with an agitator (temperature settings of 107/110/114°C with an agitator speed of 120 rpm; 114/116/120°C with an agitator speed of 120 rpm; 125/140/150°C with an agitator speed of 30 rpm respectively), is continuously charged with a feed composed of 12 parts by weight of a functionalized rubber, 55.5 parts by weight of styrene, 17.5 parts by weight of acrylonitrile and 15 parts by weight of ethyl benzene at a rate of 900 g/hr. The initiator, 1,1-di(t-butyl peroxy) cyclohexane and is added to the top of the first reactor. N-dodecylmercaptan (nDM)(chain transfer agent) is added to optimize the rubber particle sizing and the matrix molecular weight. The functionalized rubber is a styrene/butadiene rubber containing 20 weight percent styrene, functionalized with 2,2,6,6-tetramethyl-1-[1-[4-(oxiranylmethoxy)phenyl]ethoxy]-piperidine and having a

solution viscosity of 52 cps. After passing through the 3 reactors, the polymerization mixture is guided to a separation and recovery step using a preheater followed by a devolatilizing extruder. Finally the molten resin is stranded, cooled and cut in granular pellets.

Solution viscosity is measured in 5 wt. percent solution in styrene at 25°C. RPS (rubber particle size) is measured using Coulter Counter (20 μm orifice). SB (styrene-butadiene) block copolymers are produced according to the process described in US Patent 5,721,320 (Priddy).

Intrinsic Gloss is determined by 60° Gardner gloss on specimens prepared from molded samples, 30 minutes after molding, according to ISO 2813 with "Dr. Lange RB3" reflectometer.

Intrinsic gloss specimens were molded on an a DEMAG injection molding machine model D 150-452, having the following molding conditions: Barrel temperature settings of 210, 215, and 220°C; Nozzle temperature of 225°C, Mold temperature of 30°C; Injection pressure: 1500 bar; Holding pressure 50 bar; Holding time 6 seconds; Cavity switch pressure: 200 bar; Cooling time: 30 seconds; and injection speed: 10 cubic centimeters per second (cm^3/s).

The dimensions of the molded plaque are 64.2 mm x 30.3 mm x 2.6 mm. Intrinsic gloss is measured in the center of the plaque on the surface at which the pressure is measured. The materials are injected through one injected point located in the middle of the short side of the mold. During injection molding, the injection pressure switches to holding pressure when the cavity pressure reaches the pre-set value. The pressure transducer is located at a distance of 19.2 mm from the injection point.

The polishing of the mold is according to SPI-SPE1 standard of the Society of Plastic Engineers.

The following Table contains further details with respect to run conditions (different initiator and/or chain transfer agent amounts) and properties. From the Table it is obvious that the RPS is larger for Comparative Example A having a functionalized rubber with a solution viscosity greater than 50 cps than Examples 1 to 3 in the application in which the rubbers have a solution viscosity less than 50 cps, the larger particle size is reflected in a lower intrinsic gloss.

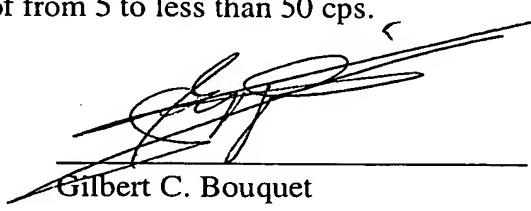
Run	Initiator top R1 (ppm)	Comparative Example A			
		nDM top R1 (ppm)	nDM top R2 (ppm)	RPS (μm)	Intr.Gloss (%)
		100	250	1000	67
1	100	250	1200	0.63	64
2	90	250	1200	0.66	66
3	90	200	1200	0.59	69
4	90	150	1600	0.75	61

Conclusions

(1) From the data presented in Figure 1, a rubber having a Mw of 3930 kg/mole would have a solution viscosity significantly lower than 5 cps.

(2) An ABS produced by the process of the invention with the exception that the solution viscosity of the functionalized rubber is outside the range of from 5 to less than 50 cps does not provide the desirable higher gloss as do ABS products made by the process of the present invention that utilize functionalized rubbers having a solution viscosity within the claimed range of from 5 to less than 50 cps.

Further affiant sayeth not.



Gilbert C. Bouquet

Sworn to and subscribed before me this 10 day of October, 2007.

XXXXXX
(Notary Public)

-(~~SEAKX~~)

*Subscribing Witness Affidavit
Execution Form attached

This Form 882 is in the ENGLISH language and has the same meaning in other languages with the same 882 number.

FORM 882

Dow Case No. 62723A

**SUBSCRIBING WITNESS
AFFIDAVIT OF EXECUTION**

I, Sultan Erikci-Yilmaz

Whose full post office address is:

Coehoornstraat 5
4551 HW Sas van Gent
The Netherlands

Make oath and say that I was personally present and did see:

Gilbert Bouquet

known or identified to me to be the individual(s) described in and who executed the foregoing document

Affidavit

and acknowledged the same as a free act and deed for uses and purposes therein expressed.

Signed at: _____ Terneuzen _____

On this 10 day of October, 2007,


Subscribing Witness

07/00

Related Proceedings Appendix

None applicable